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# Wintertime characteristics of aerosols over middle Indo-Gangetic Plain: Vertical profile, transport and radiative forcing☆

M. Kumar [a], M.P. Raju [b], R.K. Singh [c], A.K. Singh [d], R.S. Singh [e], T. Banerjee [a,]*

[a] *Institute of Environment and Sustainable Development, Banaras Hindu University, Varanasi, India*
[b] *National Centre for Medium Range Weather Forecasting, Noida, India*
[c] *International Center for Agriculture Research in the Dry Areas, New Delhi, India*
[d] *Atmospheric Research Laboratory, Department of Physics, Banaras Hindu University, Varanasi, India*
[e] *Department of Chemical Engineering and Technology, Indian Institute of Technology (BHU), Varanasi, India*

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ABSTRACT

Winter-specific characteristics of airborne particulates over middle Indo-Gangetic Plain (IGP) were evaluated in terms of aerosol chemical and micro-physical properties under three-dimensional domain. Emphases were made for the first time to identify intra-seasonal variations of aerosols sources, horizontal and vertical transport, effects of regional meteorology and estimating composite aerosol short-wave radiative forcing over an urban region (25°10′–25°19′N; 82°54′–83°4′E) at middle-IGP. Space-borne passive (Aqua and Terra MODIS, Aura OMI) and active sensor (CALIPSO-CALIOP) based observations were concurrently used with ground based aerosol mass measurement for entire winter and pre-summer months (December, 1, 2014 to March, 31, 2015). Exceptionally high aerosol mass loading was recorded for both $PM_{10}$ ($267.6 \pm 107.0\ \mu g\ m^{-3}$) and $PM_{2.5}$ ($150.2 \pm 89.4\ \mu g\ m^{-3}$) typically exceeding national standard. Aerosol type was mostly dominated by fine particulates (particulate ratio: 0.61) during pre to mid-winter episodes before being converted to mixed aerosol types (ratio: 0.41–0.53). Time series analysis of aerosols mass typically identified three dissimilar aerosol loading episodes with varying attributes, well resemble to that of previous year's observation representing its persisting nature. Black carbon ($9.4 \pm 3.7\ \mu g\ m^{-3}$) was found to constitute significant proportion of fine particulates (2–27%) with a strong diurnal profile. Secondary inorganic ions also accounted a fraction of particulates ($PM_{2.5}$: 22.5%; $PM_{10}$: 26.9%) having $SO_4^{-2}$, $NO_3^-$ and $NH_4^+$ constituting major proportion. Satellite retrieved MODIS-AOD (0.01–2.30) and fine mode fractions (FMF: 0.01–1.00) identified intra-seasonal variation with transport of aerosols from upper to middle-IGP through continental westerly. Varying statistical association of columnar and surface aerosol loading both in terms of fine (r; $PM_{2.5}$: MODIS-AOD: 0.51) and coarse particulates ($PM_{10}$: MODIS-AOD: 0.53) was found influenced by local meteorology (boundary layer and humidity) and aerosol vertical profile. A gradual increase in aerosol vertical profile (surface to 4.9 km) was evident with dominance of polluted continental, polluted dust and smoke at lower altitude. Presence of mineral dusts in higher altitude during later phase was linked with its transboundary transport, originating from western dry regions. Conclusively, winter-specific short-wave aerosol radiative forcing revealed an ATM warming effect (31–47 W $m^{-2}$) while cooling both at TOA (−20 to −32 W $m^{-2}$) and SUF (−51 to −80 W $m^{-2}$) with significant level of intra-seasonal variations in heating rates (0.86–1.32 K $day^{-1}$).



## 1. Introduction

Airborne particulates have enormous potential in modifying atmospheric heat budget either by absorbing/scattering insolation (Dey and Tripathi, 2007), altering cloud microphysical properties (Gettelman et al., 2013) and thereby, regulating regional climate and hydrological cycle (Ji et al., 2011). Characteristic features of airborne particulates are highly heterogeneous and regulated by meteorology (Banerjee et al., 2011a; Kumar et al., 2015a, 2016); regional topography (Ramanathan and Ramana, 2005) and emission sources (Badarinath et al., 2009; Banerjee et al., 2011b). Global distributions of aerosols are typically region specific due to its relatively shorter lifespan and thereby, aerosols pose a strong regional signature (Banerjee et al., 2015). Globally there are few isolated regions which are well recognized of having considerably high aerosol loading with unique region specific characteristics, notably Indo-Gangetic Plain (IGP) of South Asia, South-East Asia, tropical regions of Africa (Ji et

☆ Capsule: Intra-seasonal variations in aerosol radiative forcing was found regulated by aerosol sources, transport and aerosol constituents over middle-IGP.

* Corresponding author at: Institute of Environment and Sustainable Development, Banaras Hindu University, Varanasi 221005, India.
*E-mail addresses:* tb.iesd@bhu.ac.in, tirthankaronline@gmail.com (T. Banerjee).

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al., 2015), Eastern provinces of China, tropical region of Mexico and Central America. Among these, the IGP is often characterized with exceptionally high aerosol loading (Ramanathan and Ramana, 2005; Sen et al., 2016) with heterogeneous compositions (Dey and Tripathi, 2007; Banerjee et al., 2015; Kumar et al., 2016) typically dominated by organics (17–40%), mineral dust (11–14%) and inorganic ionic species (20–30%) (Murari et al., 2015, 2016). Presence of such diverse aerosols pose potential to severely affect regional environment (Ramanathan and Ramana, 2005) both in terms of pollution and heath (Kumar et al., 2015b), food security (Burney and Ramanathan, 2014) and other climate related services.

The main uncertainty in modeling implications of aerosols on regional climate exists on heterogeneities in aerosol chemical and micro-physical properties under three-dimensional domain. Aerosol properties strongly modify under varying emission and meteorological conditions and therefore, require extensive region specific investigation. Winter-specific aerosol loading and properties over middle-IGP is highly versatile (Sen et al., 2016) and pose significant level of uncertainties in terms of particle morphology (Murari et al., 2016), ionic composition (Satsangi et al., 2013), optical properties (Ramanathan and Ramana, 2005), vertical distribution (Kumar et al., 2015a), transport (Nair et al., 2007) and radiative forcing (Dey and Tripathi, 2007). Such uncertainties in aerosol characteristics initiate the scope of identifying the regional characteristics of winter-specific aerosols over the region.

Present submission focuses on analyzing both ground based and satellite-retrieved observations of aerosol physical, chemical and optical properties over middle-IGP. To our understanding, for the first time intra-seasonal winter-specific variations in aerosols sources, its subsequent transport, effect of regional meteorology, transformation of aerosol ionic constituents and implication of composite aerosols on shortwave radiative forcing were reported. Wintertime aerosol mass loading was initially analyzed to recognize any episodic and repetitive nature of particulate loading before being statistically assessed in terms of its association with regional meteorology. Intra-seasonal episode specific variation in aerosol ionic constituents and BC mass loading were also analyzed to recognize influences of various sources. Aerosols columnar properties were retrieved both over Indian geographical region and specifically over middle-IGP. Initially, columnar aerosols both in terms of AOD and fine mode fractions (FMF) were assessed for entire Indian geographical region to understand aerosols spatial distribution. Further, aerosols columnar properties retrieved both through satellite and ground based photometers were analyzed in respect to surface particulate (PM) mass loading. Meteorological implications in regulating AOD-PM relations over middle-IGP were also assessed. Lidar based observations of aerosol vertical profiles were concurrently discussed using particle back trajectory to realize regional and trans-boundary transport of aerosols. Conclusively, surface measured particulate and BC mass loading were used to understand intra-seasonal variation of aerosol radiative forcing under varying humidity and particulate loading scenarios. The implications of such analysis may well be useful in recognizing uncertain inferences of intra-seasonal variations of aerosols on regional climate under changing particulate emission and meteorological conditions.

## 2. Site description and meteorology

Present study has been carried out in Varanasi (25°16′N, 82°59′E, 82.2 m AMSL), a typical urban environmental setup having characteristic features ideally representing the middle-IGP (Kumar et al., 2015a). The city is surrounded by rural and semi-urban localities (Fig. 1) without any significant impact from local industrial emissions. Predominant sources of airborne particulates include fossil fuel combustion through vehicles, re-suspended dust, biomass burning and waste incineration (Murari et al., 2015). Contribution of aerosols of trans-boundary origin is also evident recurrently (Kumar et al., 2015a).

The region is climatologically affected by wide range of weather with minimum localized impacts of ocean and mountain. Additionally, relatively flat landscape simplifies the atmospheric boundary layer (ABL) over the region. The region experiences humid sub-tropical climate with clear seasonal distinction having extreme summer (March to June, 38–43 °C), wet monsoon (July to September, 80% of the annual rainfall) and cold during winter (December to February, 5–15 °C). Meteorological parameters i.e. temperature, relative humidity (RH) and wind speed (WS) were obtained from www.wunderground.com and further validated with India Meteorological Department station located inside the university campus. The average temperature was 18.6 ± 5.0 °C having minimum during the last week of December (8.9 °C) and maximum during end of March (30.0 °C). RH varied from 44% (late March) to 98% (early January) with an average of 74.9 ± 13.6%. WS was relatively calm (23% of monitoring period) during the entire period (1.3 ± 0.7 $ms^{-1}$; range: 0.4–3.6 $ms^{-1}$). However, thermal inversion, reduced convection, shallow ABL (423 ± 162 m; range: 197–932 m) and periodical dense foggy conditions additionally complicate the regional atmospheric chemistry of airborne particulates.

Intra-seasonal variability in ABL height was retrieved from National Oceanic and Atmospheric Administration, Air Resources Laboratory (NOAA-ARL), Real-time Environmental Applications and Display System (READY) website (http://www.arl.noaa.gov/ready; Draxler and Rolph, 2003).

## 3. Experimental methods

### 3.1. Surface based in-situ measurements

#### 3.1.1. Aerosols mass loading

Airborne particulates with aerodynamic diameter ($d_{ae}$) ≤10 μm ($PM_{10}$) and ≤2.5 μm ($PM_{2.5}$) were monitored for the entire period (December 1, 2014 to March 31, 2015; n = 54). Particulate monitoring was continued on each alternate days at a height of 7.0 m above ground continuously for 22 h (12:00–10.00 h). The coarse particulates ($PM_{10}$) were collected on 20.3 × 25.4 cm sized glass fiber filters (GF/A Whatman) using respirable dust sampler (APM-BL 460, Envirotech) having size selective inlet with an average flow rate of 1.2$m^3 min^{-1}$ (1.1–1.3$m^3 min^{-1}$, accuracy ±2%). Fine particulates ($PM_{2.5}$) were concurrently monitored using a fine dust sampler (APM-550, Envirotech) with a flow rate of 1 $m^3$ $h^{-1}$ (accuracy: ±2%) using polytetrafluoroethylene filter (PTFE, 47 mm diameter). All filters were desiccated for 24 h before and after particulate monitoring. Filters were pre and post-weighed after sampling using microbalance (AY220, Shimadzu) and mass concentrations were estimated gravimetrically. The exposed filters were stored under cool and dry condition (−20 °C) for further ionic analysis (Murari et al., 2015).

#### 3.1.2. Black carbon mass loading

Real time black carbon (BC) mass concentrations were monitored using a 7-channel Aethalometer (AE-42, Magee Scientific) in parallel with airborne particulates. Aethalometer measures attenuation of light (ATN) at seven channels (370, 470, 520, 590, 660, 880 and 950 nm) while attenuation corresponding to 880 nm were considered as standard BC mass loading (Bodhaine, 1995). The concept of linearity between light attenuation with the deposited BC mass on quartz filter is the key principle for estimation of BC. The details of operating principle may found in the works of Wang et al. (2012 and reference therein). An absorption efficiency of 16.6 $m^2$ $g^{-1}$ (provided by the manufacturer) was used to estimate the BC mass concentration. Shadowing effect was rectified using correction algorithm provided by Virkkula et al. (2007) and as employed by Raju et al. (2011). Aethalometer was run at an altitude of about 7.0 m above the ground using inlet tube and was operated at a constant flow rate of 5 lpm with a resolution of 5 min.

#### 3.1.3. Aerosol water-soluble ionic constituents (WSIC)

The water-soluble ionic constituents (WSIC) associated with both $PM_{10}$ and $PM_{2.5}$ were analyzed by ion chromatograph (ICS-3000, Dionex, USA). All the exposed filters corresponding to a particular loading episode